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Vegetation and Foodstuff Monitoring

S. Ring Peterson

Introduction

LLNL has a vegetation and foodstuff monitoring program to comply with Department of Energy (DOE) guidance, which states that periodic sampling and analysis of vegetation should be performed to determine if there is measurable long-term buildup of radionuclides in the terrestrial environment (U.S. Department of Energy 1991).

Tritium is the nuclide of major interest in the LLNL vegetation and foodstuff monitoring program because LLNL has historically released tritium to the air during routine operations and accidentally. Tritium moves through the food chain as tritiated water and can be rapidly assimilated into plant water and then incorporated into the organic matter of plants through photosynthesis. It can contribute to human radiation dose if it is inhaled or if vegetables or meats from animals having eaten tritiated vegetation are ingested. Although other radionuclides are released from LLNL and tritium does not build up over time, our assessments show that only tritium can be present in vegetation in detectable concentrations.

LLNL has been monitoring tritium in vegetation to some extent since 1966 and has performed vegetation sampling in the vicinity of the Livermore site and Site 300 as part of a continuing monitoring program since 1972. The monitoring program is designed to measure changes in environmental levels of radioactivity, to evaluate increases in radioactivity that might have resulted from LLNL operations, and to calculate potential human doses from radionuclides in the food chain. In 1977, LLNL added wine to the LLNL monitoring program.

During 1998, LLNL collected and analyzed samples of herbaceous vegetation and wine. Potential human doses from these foodstuffs were calculated using the monitoring data and dose models presented in Appendix A. In addition, as part of a continuing study, LLNL determined potential dose from inhalation, skin absorption, and ingestion to the maximally exposed individual from a pine tree on site. Because the roots grow in tritium-contaminated water, tritium is lost to the atmosphere via evapotranspiration, and the pine tree can be modeled as a diffuse source of tritium. Dose was calculated using the Environmental Protection Agency (EPA) model, CAP88-PC.

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Methods

Our methods for monitoring vegetation and wine are presented in the following sections.

Vegetation

LLNL collects vegetation samples, usually annual grasses, quarterly from fixed locations in the Livermore Valley, San Joaquin County, and Site 300, and then analyzes them for tritium.

Location maps are provided in **Figures 11-1** and **11-2**. These locations were selected so samples would represent vegetation from: (1) locations near LLNL that can be affected by LLNL operations, (2) background locations where vegetation is similar to that

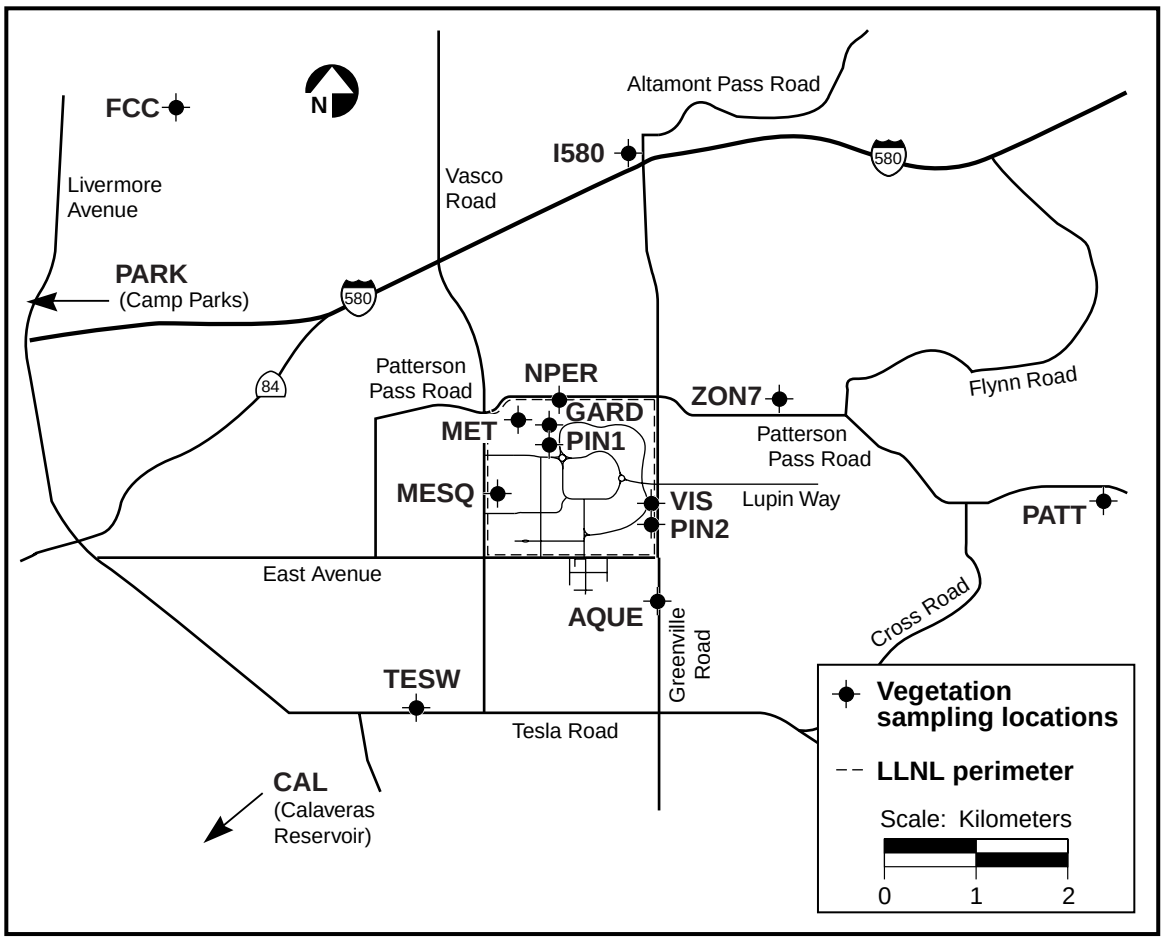

Figure 11-1. Livermore Valley vegetation sampling locations, 1998.

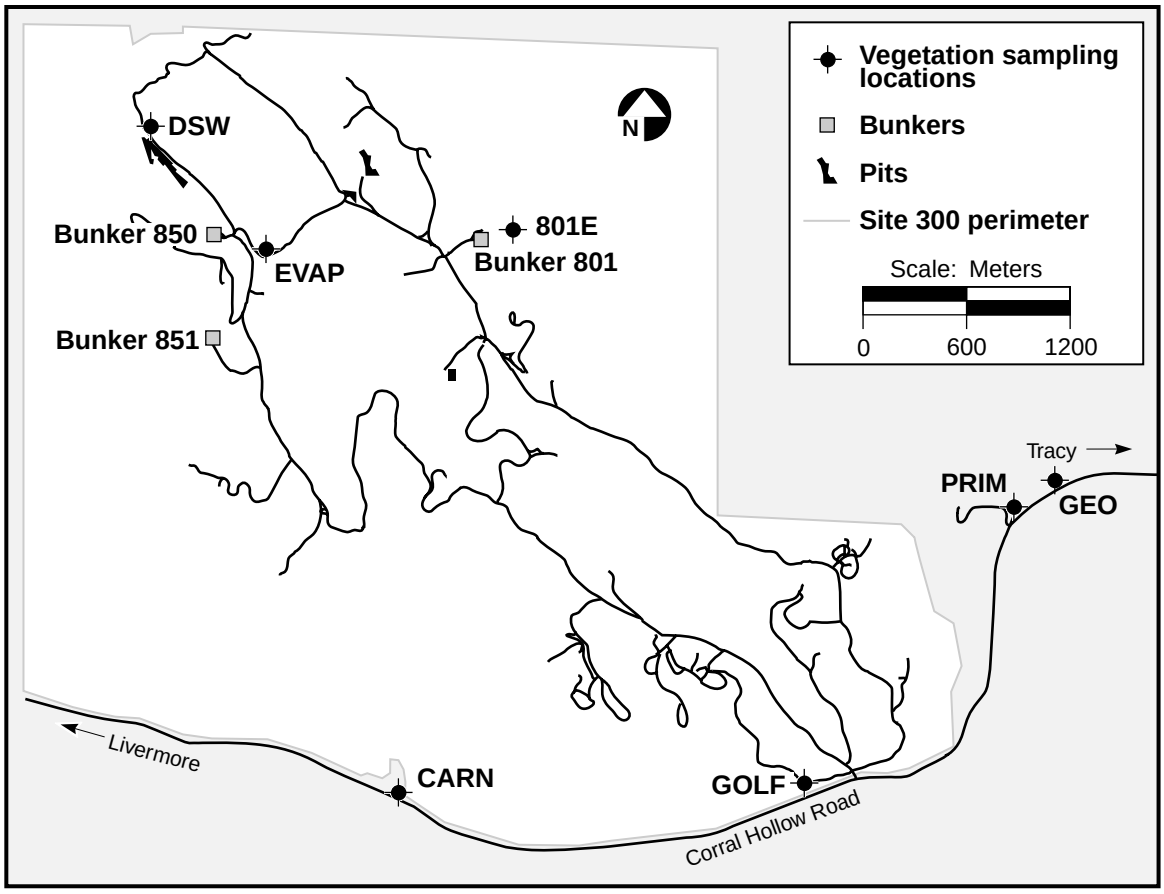

Figure 11-2. Site 300 vegetation sampling locations, 1998.

growing near LLNL but is unlikely to be affected by LLNL operations, and (3) areas of known or suspected LLNL-induced contamination. Sampling locations for 1998 were the same as in 1997, although sampling in 1998 at PIN1 and PIN2 was performed quarterly rather than monthly, as in 1997.

Vegetation samples are frozen when collected. When processed at the analytical laboratory, the samples are freeze-dried to collect the free water in the plant, and the tritium content of this water is determined by scintillation counting.

All vegetation sampling is conducted according to written and approved standardized procedures (Tate et al. 1995). Approximately 10% of the sites are sampled in duplicate to comply with quality assurance protocols.

Wine

Wine is the most important agricultural product in the Livermore Valley, representing an approximately $80-million annual industry. Although the tritium concentrations in all wines are low, the data indicate that Livermore Valley wines contain statistically more tritium than do other California counterparts.

Three types of wine samples were collected and analyzed for tritium concentrations: wine produced from grapes grown in the Livermore Valley, wine produced from grapes grown in California outside the Livermore Valley, and wine produced from grapes grown in Europe (France, Germany, and Italy).

The wines were purchased from local retailers to represent what the general public could buy during 1998 (Tate et al. 1995). Sample tritium concentrations are determined by helium-3 mass spectrometry. The resulting analytical data can be used to estimate the potential tritium dose received by consumers during the year of purchase. However, since wines purchased in 1998 are from grapes harvested in 1996 and 1997, the 1998 sampling data cannot be used to indicate how LLNL's operations affected wines produced from grapes grown in 1998. To analyze trends and help determine the impact of LLNL operations on tritium in wine for the year the grapes were harvested, LLNL corrects the wine sample concentrations for the radiological decay that has taken place between approximate date of harvest and the date of analysis in the laboratory. Comparisons can then be made of wine concentrations that represent the year the grapes were exposed to the tritium.

Approximately 10% of the total complement of wines was sampled in duplicate, 30% of all the samples were analyzed multiple times, and traceable standards were evaluated to comply with quality assurance protocols.

Results

The results of vegetation and wine monitoring for the Livermore site and Site 300 are presented in the following sections.

Livermore Site

Vegetation

The Livermore Valley vegetation locations are put into four groups for statistical evaluation:

- Near—locations at or within 1 km of the Livermore site perimeter. Near locations include AQUE, GARD, MESQ, NPER, MET, PIN2, and VIS.
- Intermediate—locations in the Livermore Valley 1–5 km from the Livermore site perimeter that are often downwind and thus potentially under the influence of tritium releases at the site. The intermediate locations are I580, PATT, TESW, and ZON7.
- Far—locations unlikely to be affected by LLNL operations. One background location (CAL) is more than 25 km away. The other two (FCC and PARK), although in the Livermore Valley, are unlikely to be affected by LLNL operations because they are more than 5 km from the Livermore site and generally upwind.
- PIN1—location of a pine tree rooted in an area of known tritium contamination on site. Locations PIN1 and PIN2 (a pine tree not rooted in an area of known tritium contamination) were studied monthly during 1997 and quarterly during 1998

Table 11-1 shows summary tritium data for vegetation collected for the LLNL vegetation monitoring program in 1998 (the individual sampling values are presented in the Data Supplement of this report). **Figure 11-3** shows the 1998 medians of the tritium concentrations for Near, Intermediate, and Far Livermore locations as well as historic median concentrations from 1971 to 1997.

In 1998, at our request, the analytical laboratory began reporting measured tritium activities, even if they were below the detection limit. This change only affected non-detected data. As a result, some numbers previously reported as “less than” values will now be negative. Since a negative median cannot be graphed on a logarithmic scale (e.g., **Figure 11-3**), the lowest positive measured concentration has been graphed instead. For the Far location, this new treatment of data has resulted in a drop in concentration between 1997 and 1998 of about a factor of 10, but it is highly likely that there is no difference between 1998 and recent preceding years.

The data for tritium in vegetation were compared using the Tukey-Kramer honestly significant difference (HSD) test. As in 1997, the evaluation of the 1998 data show a significant difference between the Near group and the other two groups; that is, the Near values are significantly higher than the Intermediate and Far values, but the Intermediate values are not significantly different from the Far values. The highest median tritium results for individual vegetation sampling locations were found at AQUE and VIS, which are located downwind of the Livermore site and historically have exhibited higher values than other locations.

Table 11-1. Tritium in vegetation (in Bq/L), 1998

Location[a]	Detection Frequency[b]	Median	Interquartile Range	Maximum	Dose (μSv/y)[c]	
					Median	Maximum
Near Livermore site[d]	23/27	4.4	4.1	22	0.021	0.10
Livermore site PIN1[e]	4/4	37	41	110	1.9×10^{-6}[f]	5.3×10^{-6}[f]
Intermediate distance from Livermore site	7/16	1.0	3.3	6.0	0.0050	0.029
Far from Livermore site	3/12	–0.29	1.8	3.6	—[g]	0.017
Location DSW at Site 300[e]	4/4	390	1200	3300	1.9	16
Location EVAP at Site 300[e]	2/4	4.7	130	510	0.023	2.5
All other locations at Site 300	0/20	–0.28	0.65	0.80	—[g]	0.0039

Note: Radioactivity is reported as the measured concentration and an uncertainty (±2σ counting error). If the uncertainty is greater than the concentration, the result is considered to be a nondetection.

a See Figures 11-1 and 11-2 for sampling locations.

b Detection frequency means the fraction of samples taken having a measured value above the detection limit.

c Dose is calculated based on conservative assumptions that an adult's diet is exclusively vegetables with this tritium concentration, and that meat and milk are derived from livestock fed on grasses with the same concentration of tritium. See Appendix A, Methods of Dose Calculations.

d Includes PIN2; free-water concentrations are similar among plant types.

e Sampling location in known area of contamination.

f For this dose calculation, PIN1 is treated as a diffuse source of tritium (since pine needles are not eaten by human beings). Dose, calculated using CAP88-PC, is to the maximally exposed individual.

g Dose is not calculated when the median concentration is negative.

In 1997, PIN1, a pine tree growing in a known area of contamination, was monitored on a monthly basis to estimate emissions for compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs). In 1998, the tree sampling was coordinated with the quarterly vegetation sampling, and NESHAPs calculations to the sitewide maximally exposed individual (SW-MEI) for 1998 are based on the observations from the quarterly samples. To assess the contribution of soil water tritium to

PIN1, LLNL also sampled a second tree that is not in tritium-contaminated soil (PIN2). Concentrations of tritium in PIN2, like all in other vegetation sampled near the Livermore site, are from air and soil water in equilibrium with air. Concentrations from PIN2 are statistically higher than other Near locations (at the 5% significance level). This may be because, compared with most of the other sampling locations, PIN2 is both closer to the main source of tritium and is in a direction towards which the wind blows frequently. It may also be caused by exposure of the needles to slightly higher air concentrations with height above ground-level.

Wine

The results from the 1998 wine tritium analyses are shown in **Table 11-2**. Tritium concentrations are within the range of those reported in previous years and remain low in wines from all areas.

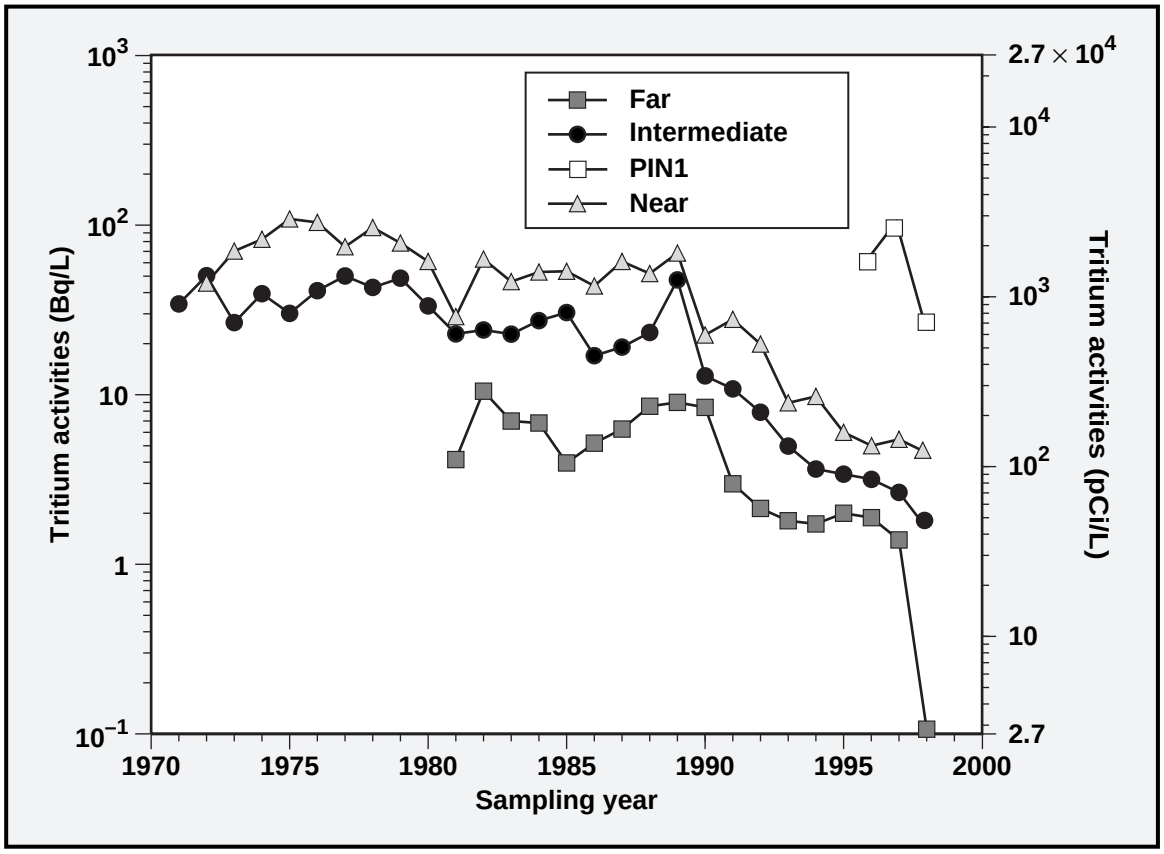

Figure 11-3. Median tritium activities in Livermore Valley vegetation samples, 1971 to 1998.

Table 11-2. Tritium in retail wine (in Bq/L), 1998.[a]

Region	Detection[b] frequency	Median	Interquartile range	Mean	Maximum	Dose[c] μSv/y
Livermore Valley	12/12	2.8	2.2	3.2	8.2	0.0028
California	6/6	0.40	0.10	0.42	0.58	0.00037
Europe	4/4	1.6	0.24	1.5	1.7	0.0013

a Wines from a variety of vintages were purchased and analyzed during 1998. The concentrations shown are those at the time the bottle was opened.

b Detection frequency is the number of samples taken with results above the detection limit divided by the number of samples.

c This dose is calculated from the assumption of drinking 52 L wine/year and using the mean concentration of sampled wines.

The data for the 1998 sampling year were analyzed using analysis of variance (ANOVA). The statistical analyses show that the mean tritium concentration of the twelve Livermore wines sampled is statistically greater than that of the six California wines sampled. The statistical analyses also indicate that there is no significant difference between the mean tritium values of the European (n = 4) and California wines sampled or between the Livermore and European wines. Multiple comparison tests indicate that the mean levels of the 1998 sampling year data from all areas are not significantly different from those reported for the 1995, 1996 and 1997 sampling years despite the visually apparent downward trend in California wines in **Figure 11-4**.

Figures 11-4 and **11-5**, which show historic means by sampling year and by vintage year, for 1998 look quite different from similar figures in previous years because concentrations have been plotted as logarithms so that differences in low concentrations can be distinguished easily.

Helium-3 mass spectrometry has been used since 1991 to analyze the wine data. Since results from different methods of analysis may not be strictly comparable, only wines measured with ^{3}He mass spectrometry have been compared for trends in decay-corrected concentrations, grouped by vintage year. Wines measured during this period include 1988–1997 vintages for California and Europe and 1983–1997 vintages for the Livermore Valley. Regression analysis shows tritium concentrations in California and European wines decreasing very slightly, while the tritium concentrations in Livermore wines are decreasing more sharply. However, none of these regression lines is statistically significant ($p < 5\%$). In support of the regression analyses, the ANOVA results show some statistical differences between earlier (before 1988) and later vintage wines for Livermore but not for California or Europe.

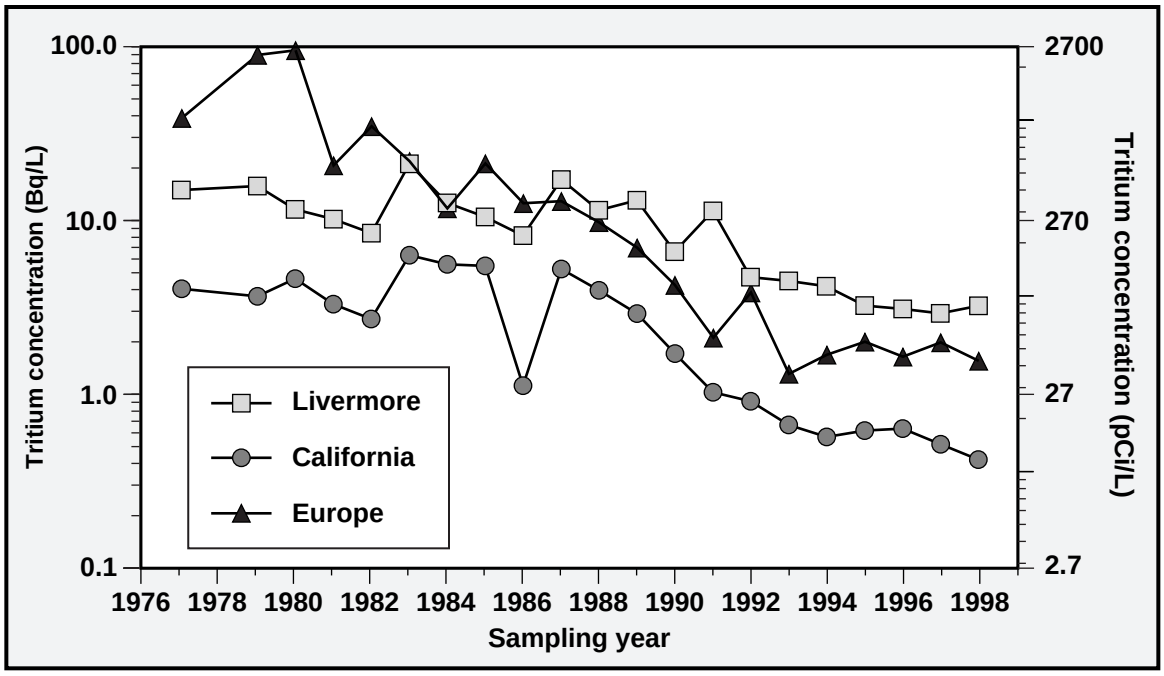

Figure 11-4. Mean tritium in retail wines, 1977 to 1998, plotted as measured for each sampling year.

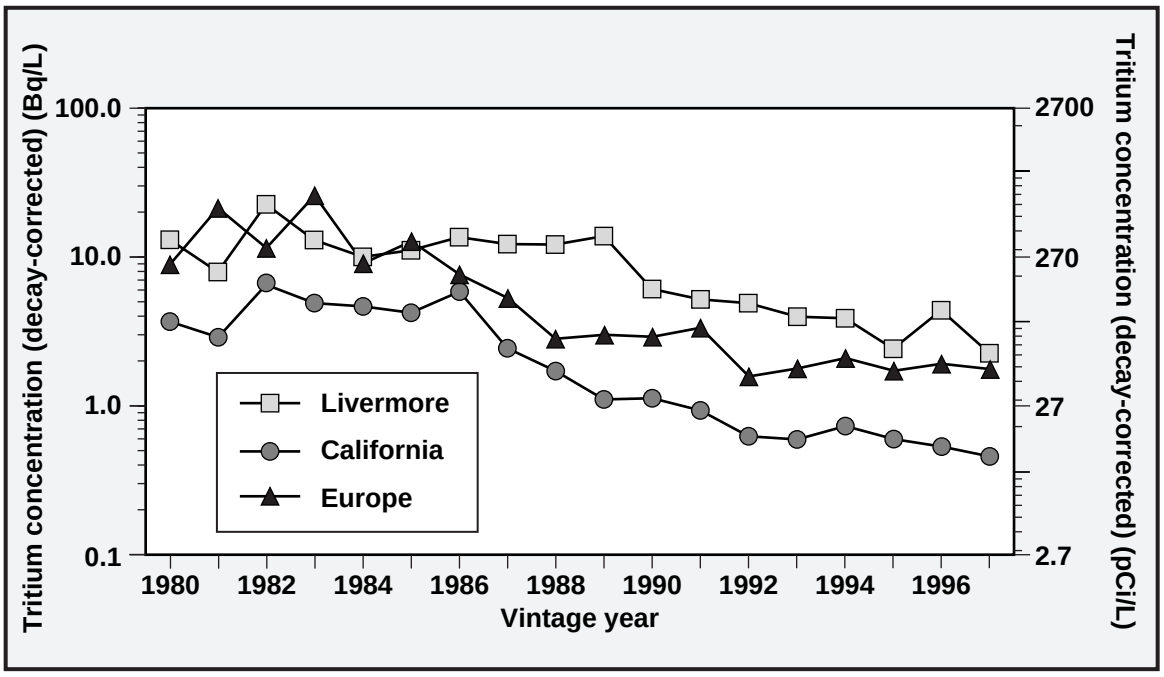

Figure 11-5. Mean tritium concentrations in retail wines decay-corrected from the sampling year to the vintage year.

When all wines for 1988–1997, decay-corrected to vintage year, are compared, the concentrations in Livermore wines show higher tritium concentrations than do Californian or European wines, which are indistinguishable statistically (at the 5% significance level) for that time period.

Site 300

Vegetation

Table 11-1 shows summary tritium data for vegetation collected at Site 300 during 1998. Historic values for tritium at Site 300 sampling locations are shown in **Figure 11-6**. Of the seven sampling locations at Site 300, five yielded results at or near the detection limits. Two locations, EVAP and DSW, yielded results above background levels. The extremely low concentration for all other locations is due to graphing the lowest positive result since the median is negative and the scale of the graph is logarithmic.

The highest tritium result for a single vegetation sample occurred at location DSW (see **Table 11-1**) where tritium has been observed in the vegetation since 1971. In 1996 and 1997 plants with relatively long taproots were collected (*Urtica dioica* and *Datura wrightii*, respectively); however, in 1998 a grass with seemingly shallow roots had the highest observed tritium concentration (3300 Bq/L). This sampling location is adjacent to a landfill that contains debris contaminated with tritium from past experiments. The landfill area is currently being investigated under the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) for tritium contamination of ground water. Tritium in soil and ground water has been studied as part of LLNL's Environmental Restoration Program (Lamarre 1989a, b, and c; Taffet et al. 1989a and b; Taffet et al. 1991; Carlsen 1991a and b; and Webster-Scholten 1994). The tritium values in vegetation are also above background levels at location EVAP, which is near a spring where ground water flows near the surface and evaporates. The ground water in this area is contaminated with tritium that comes from three sources: Pit 3, Pit 5, and the firing table at Building 850 (see discussion of Wells NC7-61 and NC7-69 in Chapter 9, Ground Water Monitoring). Evaluation of the 1998 data for Site 300 using the Tukey-Kramer HSD test yielded no significant differences among the various sampling locations; this is a result of the high variability of the data and the low number of data points. Although it would be expected that combining data from earlier years (as was done in 1997) would reveal differences between at least locations DSW and EVAP compared with the others, this cannot be done because the 1998 results, with actual measured values, cannot be compared with "less than" detection limits used in previous years.

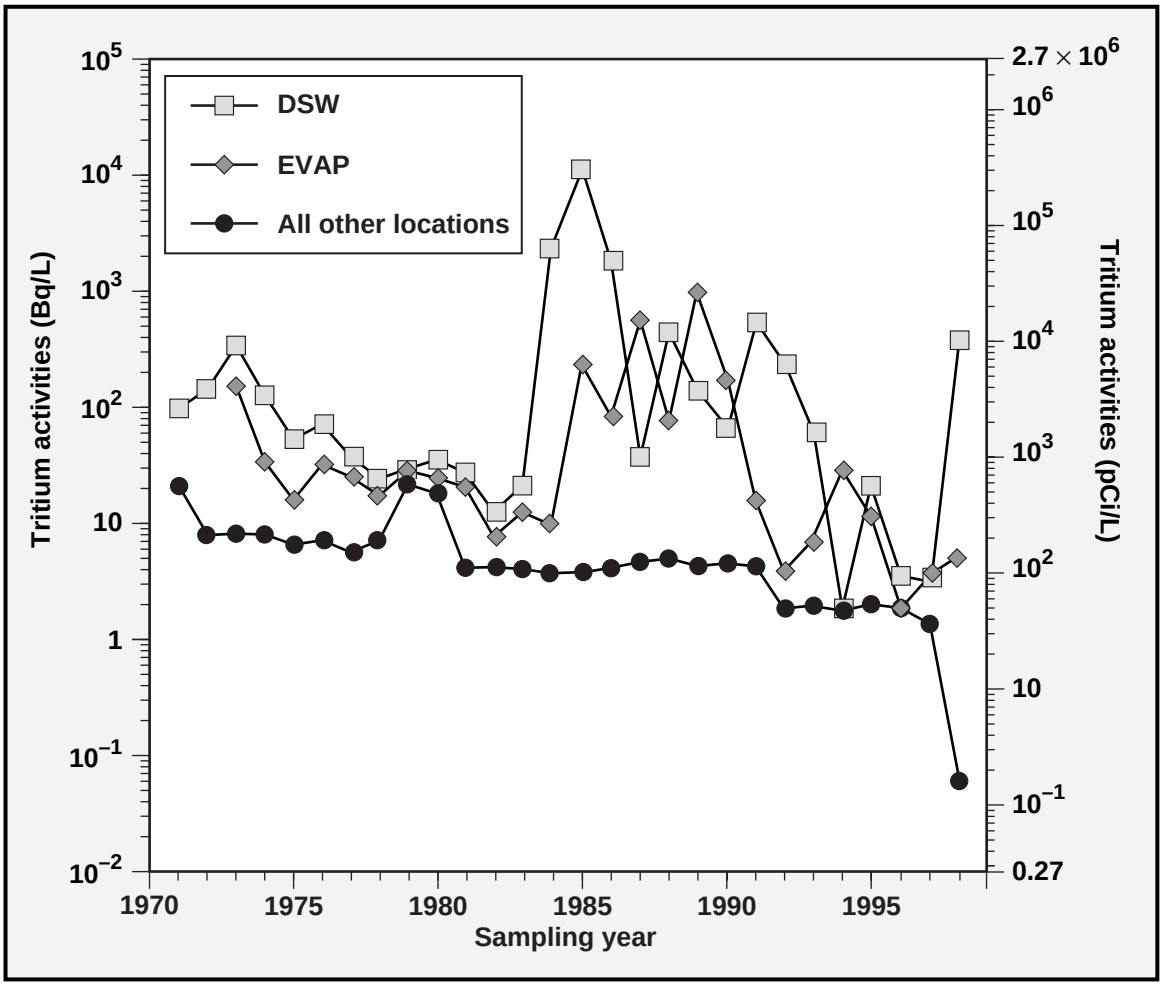

Figure 11-6. Median tritium activities in vegetation at Site 300 sampling locations, 1971 to 1998. When the median is negative (e.g., all other locations), the lowest positive concentration has been substituted.

Environmental Impact

The environmental impacts of LLNL operations on vegetation and wine monitoring are small and are presented below for the Livermore site and Site 300.

Livermore Site

LLNL impacts on vegetation in the Livermore Valley remained minimal in 1998. The effective dose equivalents shown in **Table 11-1** were derived using the dose conversion factors provided by DOE (U.S. Department of Energy 1988) and the dose pathway model from U.S. Nuclear Regulatory Commission Regulatory Guide 1.109 (U.S. Nuclear Regulatory Commission 1977). Appendix A provides a detailed discussion of dose calculation methods. The dose from tritium in vegetation is based on the conservative assumptions that an adult's diet consists exclusively of leafy vegetables with the measured tritium concentrations and meat and milk from livestock fed on grasses with the same concentrations. Most vegetables consumed by an adult will contain tritium at lower levels than those reported because they will have been imported from other areas. Similarly, tritium concentrations in food consumed by local livestock will be at or below the concentrations in vegetation measured at the Intermediate and Far locations. Nevertheless, based on these extremely conservative assumptions, the maximum potential dose from ingestion of affected vegetation for 1998 for the Livermore site is 0.10 μSv (0.010 mrem).

Doses are calculated based on measured tritium in plant water; contribution of organically bound tritium (OBT) is not included. Dose conversion factors of 1.8×10^{-11} Sv / Bq for tritium in the plant or animal water (HTO) and 4.2×10^{-11} Sv / Bq for OBT have been published by the International Commission on Radiological Protection (International Commission on Radiological Protection 1996). These show the relative importance of ingested HTO and OBT to dose. In vegetables, the dose from HTO is greater because the fraction of the plant that is organic matter is quite small (10–25%). For example, about 10% of the ingestion dose from leafy vegetables (about 10% dry matter) is from OBT. OBT becomes increasingly important when the fraction of dry matter increases. Pork, for example, has a dry matter content of about 30–50% (Ciba-Geigy Ltd. 1981), and the resulting ingestion dose from pork will be about half from OBT and half from HTO. The OBT in grain, which is 88% dry matter, will contribute nearly 90% of the total grain ingestion dose. Given the differences in OBT dose contribution from different foods, the importance of OBT to ingestion dose depends on what quantities of what kinds of foods are consumed. An extremely unlikely diet very high in OBT would, at most, give an OBT contribution to dose equal to that of HTO. Thus, conservatively, the maximum total tritium dose from ingestion of vegetables and foodstuffs from the Livermore valley could be 0.20 μSv (0.020 mrem), well below any level of concern.

The dose values for PIN1 shown in **Table 11-1** are calculated in a different manner than those for edible vegetation because it is unreasonable to assume that any person is directly ingesting pine needles. The pine tree is treated as a diffuse source of tritium to the atmosphere via the contaminated transpirational stream. LLNL uses an estimated tritium transpiration rate from the tree as input data to the U.S. EPA regulatory model CAP88-PC. LLNL models air dispersion of the transpired tritium and calculates a resulting dose from inhalation, skin absorption, and potential ingestion from air concentrations at the location of the maximally exposed individual. This total dose is based on the conservative assumptions that 100% of the individual's time is spent at this location and that his/her diet consists exclusively of vegetables with the measured tritium concentration and meat from livestock fed on grasses with the same concentration. The resulting dose for PIN1 of 5.3×10^{-6} μSv (5.3×10^{-7} mrem) is considerably lower than ingestion doses calculated directly from concentrations in vegetation because the tree is only an indirect source of air/vegetation contamination.

No health standards exist for radionuclides in wine. However, all the wine tritium levels were far below drinking water standards. In fact, even the highest detected Livermore Valley value (8.2 Bq/L or 222 pCi/L) represents only 1.1% of the California drinking water standard (740 Bq/L or 20,000 pCi/L). Doses from wine consumption can be calculated according to methods for water ingestion, described in Appendix A.

Based on the extremely conservative assumption that wine is consumed at the same rate as water (730 L/year or 2 L/day), the annual dose that corresponds to the highest detected 1998 Livermore Valley tritium value in wine is 0.10 μSv (0.010 mrem). Assuming a more realistic wine consumption (52 L/year or 1 L/week of wine) and the mean tritium values from the three sampling areas, annual doses from Livermore, European, and California wines would be 0.0028 μSv (0.00028 mrem), 0.0013 μSv (0.00013 mrem), and 0.00037 μSv (0.000037 mrem), respectively.

The potential ingestion dose from all foodstuffs grown near the Livermore site is realistically well below 0.3 μSv (0.03 mrem). This is more than a factor of 10,000 lower than an annual background dose (~3000 μSv or 300 mrem) and a factor of 330 lower than the dose from a typical chest x-ray (100 μSv or 10 mrem) (Shleien and Terpilak 1984). Therefore, although tritium levels are elevated slightly near the Livermore site, doses from tritium ingestion are negligible.

Site 300

In general, LLNL impacts on tritium concentrations in vegetation at Site 300 for 1998 were insignificant. With the exception of vegetation from previously identified sites of contamination, the tritium levels at Site 300 were at or near the limits of detection and comparable to those observed in previous years. The areas where tritium is known to be present in the subsurface soil are well delineated and localized.

The calculated maximum potential annual ingestion dose from vegetation at sampling location DSW, based on the maximum value of 3300 Bq/L (89,000 pCi/L), is 16 μSv (1.6 mrem). This dose, based on conservative modeling assumptions described above, is theoretical, since vegetation at Site 300 is not ingested either by people or by livestock. In comparison, the potential annual dose (also theoretical) from vegetation at all other locations at Site 300 could not be calculated since the median concentration is negative and doses cannot be negative.